Magnetic Ordering Temperatures Across the $RNiAl_4$ Series

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A compilation of the known magnetic ordering temperatures for the $RNiAl_4$ series of compounds is presented together with predictions for some others yet to be determined. New magnetisation data for $DyNiAl_4$ show antiferromagnetic order at 21 K in agreement with prediction. However, contrary to prediction, $HoNiAl_4$ does not show order down to 8K.

1. Introduction

The rare earth intermetallic series $RNiAl_4$ (R = rare earth) forms with the orthorhombic (space group #63, Cmcm) $YNiAl_4$-type structure in which the R and Ni atoms each occupy single crystallographic sites. The Ni sites are non-magnetic but the systems exhibit some intriguing magnetic properties due entirely to rare earth magnetism. For the compounds with R = Pr, Tb and Gd it is known that there are at least two magnetic phase transitions as a function of decreasing temperature, an intermediate incommensurate phase and a low temperature linear antiferromagnetic phase (e.g. [1-3] and references therein). In addition, the low temperature magnetisation of $TbNiAl_4$ reveals at least two transitions as a function of applied magnetic field [4]. The choice of rare earth is crucial in determining the magnetic anisotropy. For example, the Kramer's ion Nd in $NdNiAl_4$ orders along the b-axis [1,5], perpendicular to that of neighbouring non-Kramer's $PrNiAl_4$ which orders along the a-axis [1,6]. Non-Kramer's $TbNiAl_4$ also shows a-axis ordering. Furthermore $NdNiAl_4$ and $ErNiAl_4$, involving Kramer's ions, do not appear to show any obvious signs of a second magnetic phase transition, at least down to 2 K [7].

A range of techniques have been used in the study of the $RNiAl_4$ series, including bulk techniques such as heat capacity, magnetisation, susceptibility and resistivity, as well as the microscopic techniques of neutron scattering, low temperature nuclear orientation and Mössbauer spectroscopy (see references). Here we explore the emerging trends in the temperature dependence of magnetic ordering across the series. Predictions of the ordering temperatures for other $RNiAl_4$ compounds are made, and tested by way of new magnetisation measurements for $DyNiAl_4$ and $HoNiAl_4$.

2. Experimental details

The $RNiAl_4$ compounds used in this work were synthesised by repeated argon arc melting of stoichiometric amounts of 99.9% pure rare earth plus 99.99% pure Ni and Al. The resulting ingots were wrapped in tantalum foil and annealed under vacuum at 1100 °C for several days. Next the materials were checked for impurity phases using powder x-ray diffraction (XRD) and this was followed by further annealing as required. Powder was pressed into pellets for the magnetisation measurements. These measurements were carried out using a vibrating sample magnetometer with an applied magnetic field of 0.1 T. Temperatures down to approximately 8 K were achieved using a cryocooler.

3. Compilation of ordering temperatures and predictions

Experimentally measured ordering temperatures T_N (as well as second transition temperature $T_{N'}$ where it is known to occur) for some $RNiAl_4$ are listed in table 1, together with estimates of T_N for some other R. The estimates of T_N were made using the slope of the line fitted to Fig. 1, which is a plot of T_N verses the R^{3+} ground state spin component $|g_J - 1|J$. For strongly spin-orbit coupled ions and a purely exchange driven magnetic order, T_N is expected be proportional to spin only ($|g_J - 1|J$ is the projection of the total angular momentum J of the ion onto the spin S. See for example [9]).

Table 1. Néel temperatures for selected $RNiAl_4$ compounds. Those entries with a reference are experimental values. The bracketed values are estimates based on Fig. 1.

R	$\|g_J-1\|J$	T_N [K]	$T_{N'}$ [K]	Ref.
Pr	0.8	8.1	6.9	[1]
Nd	1.23	9.3	-	[1]
Sm	1.79	(15)		
Gd	3.5	24.7	20.8	[2]
Tb	3.0	34.0	28.0	[3]
Dy	2.5	(21)		
Ho	2.0	(16.5)		
Er	1.5	5.8	-	[7]
Tm	1.0	5	-	[8]

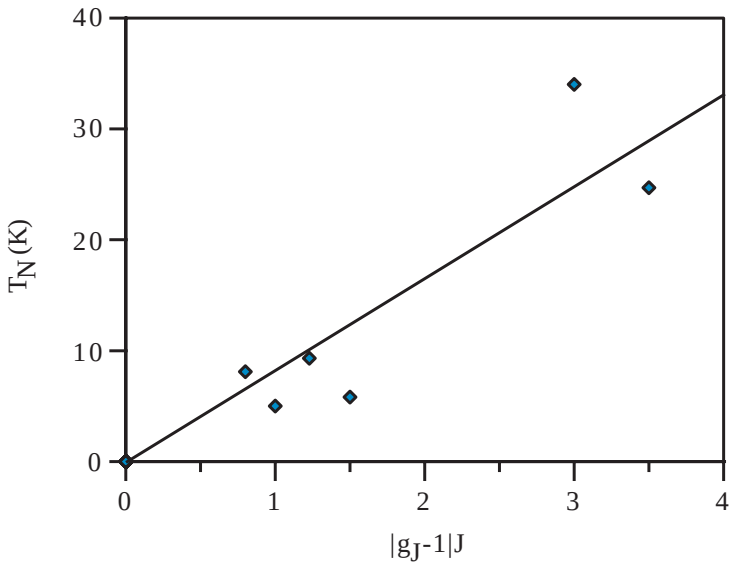

Fig. 1. Experimental Néel temperatures for various $RNiAl_4$ plotted against the spin coefficient, $|g_J - 1|J$. The straight line fit is used to predict the bracketed values of table 1.

4. Experimental results

The magnetisation data recorded for $DyNiAl_4$ and $HoNiAl_4$ are shown in Figs. 2 and 3, respectively. From Fig.2, a broad roll-over to antiferromagnetic order, centred at ~21 K, is apparent for $DyNiAl_4$. In contrast, from the data $HoNiAl_4$ (Fig. 3) there is no sign of a magnetic transition down to 8 K.

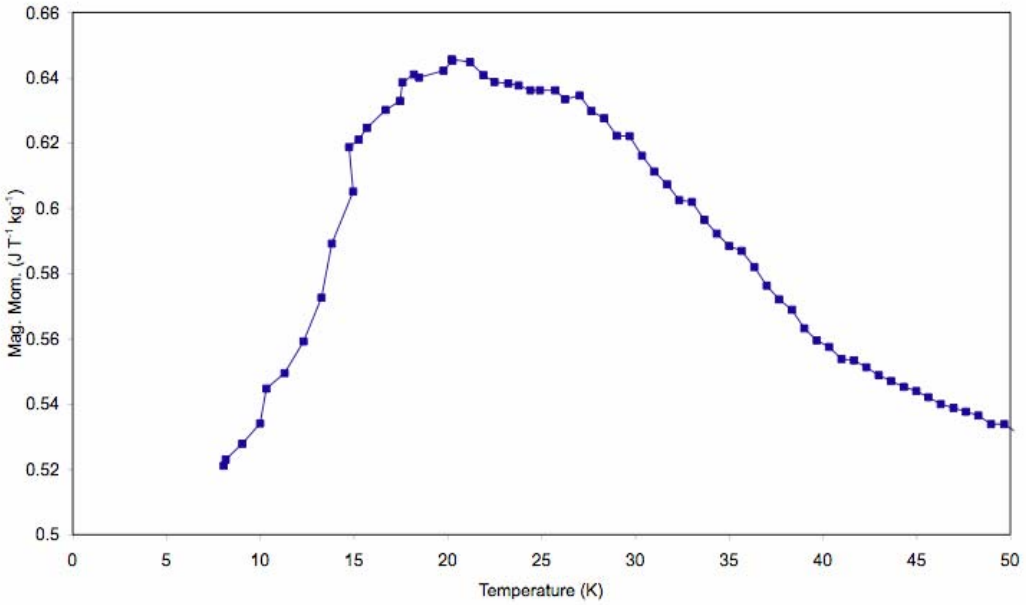

Fig. 2. Magnetisation data for polycrystalline $DyNiAl_4$.

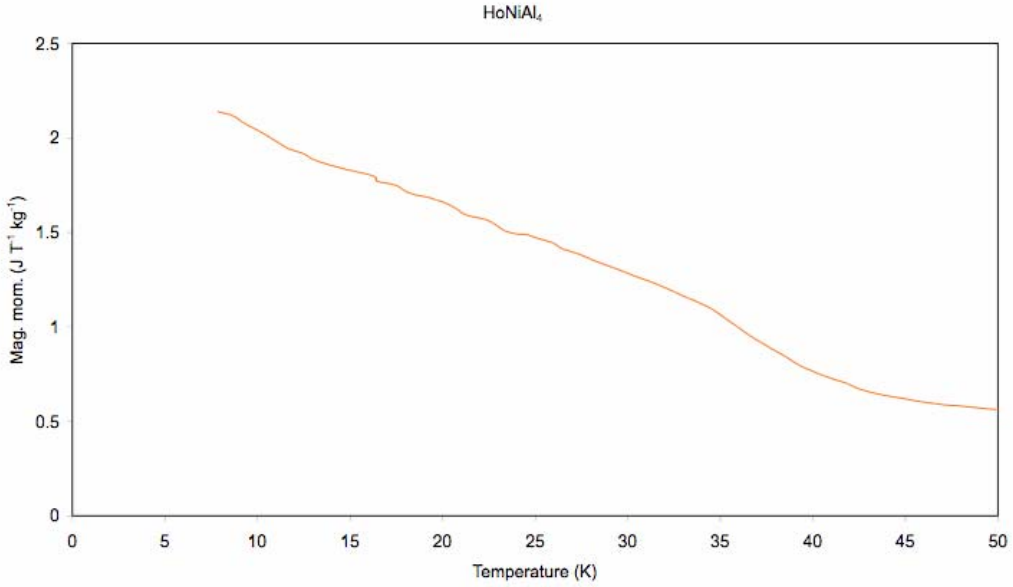

Fig. 3. Magnetisation data for polycrystalline $HoNiAl_4$.

5. Discussion

The prediction of magnetic ordering temperature based on a linear relationship with S_z across the series proved to be good for $DyNiAl_4$, with both experiment and prediction giving T_N ~21 K. However, the prediction does not work for $HoNiAl_4$, where possibly crystal field effects are more important. Additionally, we note that the broad nature of the peak in the $DyNiAl_4$ magnetisation could reflect the presence of two magnetic phase transitions. This could be an interesting result, as the first Kramer's ion in the series to show two transitions. Of course, a heat capacity and/or a single crystal magnetic measurement will be needed to confirm this speculation.

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